NAA-SR-211

Subject Category: METALLURGY AND CERAMICS

UNITED STATES ATOMIC ENERGY COMMISSION

CHANGE IN MAGNETIC SUSCEPTIBILITY OF IRRADIATED GRAPHITE DURING PULSE ANNEALING

By
J. D. McClelland

December 18, 1952

North American Aviation, Inc.
Downey, California

Technical Information Service, Oak Ridge, Tennessee

Date Declassified: December 5, 1955.

Printed in USA, Price 15 cents. Available from the Office of Technical Services, Department of Commerce, Washington 25, D. C.

CHANGE IN MAGNETIC SUSCEPTIBILITY OF IRRADIATED GRAPHITE DURING PULSE ANNEALING

WRITTEN BY:

J. D. McCLELLAND

WORK DONE BY:

J. D. Mc CLELLAND
J. J. DONOGHUE

ATOMIC ENERGY RESEARCH DEPARTMENT
NORTH AMERICAN AVIATION, INC.
P. O. BOX 309 DOWNEY, CALIFORNIA

SUBMITTED: DECEMBER 18, 1952
ISSUE DATE
FEBRUARY 24, 1953

CONTRACT AT 11-1-GEN-8

ABSTRACT

Pulse annealing studies are presented of the magnetic susceptibility of AGOT-KC graphite with irradiations of 12.5, 48, 146, 460 and 1534 mwd/ct in the Hanford "B" test hole. The samples were pulsed stepwise in steps of approximately 100° C to a temperature of 2180° C where the samples showed complete recovery to their pre-irradiation value. The curves show two active annealing regions, the first between 100° C and 500° C and the second between 1100° C and 1500° C. The data are in good agreement with theory.

This report is based upon studies conducted for the Atomic Energy Commission under Contract AT-11-1-GEN-8.

I. INTRODUCTION

The change in the diamagnetic susceptibility of AGOT-KC graphite due to radiation damage has been described in a previous report.[1] It is the purpose of the present report to describe the completion of the experimental study of this graphite by investigation of the subsequent removal of radiation damage by pulse annealing. Five irradiated graphites with exposures ranging from 12.5 mwd/ct to 1534 mwd/ct in the Hanford reactors have been studied up to an annealing temperature of 2180° C, where complete recovery of the susceptibility is indicated.

Graphite is a semi-conductor with a zero energy gap between bands. The susceptibility is believed to be primarily an electronic phenomena in which the Peierls diamagnetism predominates over all other contributions, yielding the characteristically high values reported previously.[1] If this is true, it is the only electronic property which will not depend at least implicitly on the transport characteristics of the conduction electrons. The behavior of the susceptibility during pulse annealing can therefore be expected to yield directly information regarding the change in the number of carriers during recovery from radiation damage.

II. EXPERIMENTAL METHODS AND DATA

A. Susceptibility Measurements

A Faraday non-uniform field method is used in which the force on the sample is proportional to the product of the magnetic field and its rate of change in the direction of the force. Magnet pole pieces are used whose contours generate a constant force field within a volume larger than the sample. The force exerted on a sample in the magnetic field is measured using an electrodynamical balance, in which the mechanical force is opposed by the force on a current carrying coil attached to the beam of the balance. This current is measured on a multi-range milliammeter with an accuracy of 0.1 per cent full scale. Changes in forces of the order of 0.01 dynes can be detected.

In strong fields the presence of ferromagnetic impurities will cause an appreciable change in the force exerted on the sample. This unwanted contribution to the susceptibility can be eliminated by its dependence upon the magnetic field strength; the relation between the apparent susceptibility and the true susceptibility being given by [2]

$$\chi_{(\text{apparent})} = \chi_{(\text{true})} + \frac{C\sigma}{H}$$

where C is the mass concentration of the ferromagnetic impurity and σ is the specific saturation intensity. By measuring the susceptibility at four widely separated field strengths and plotting these values against $\frac{1}{H}$, the resulting straight line can be extrapolated to yield $\chi_{(\text{true})}$. Measurements were made at field strengths ranging from 8500 oersteds to 16,000 oersteds. The apparatus has been calibrated to yield absolute measurements of the susceptibility, and this calibration is checked periodically by measuring the specific susceptibility of a sample of spectroscopically pure gold. This value is found to be 0.143×10^{-6} cgs which is slightly less than the accepted value of 0.145×10^{-6} cgs. The precision assigned to relative measurements is 0.5 per cent. In NAA-SR-153, absolute values for graphite were reported which are slightly higher than those presented in this paper. A more precise determination of the magnetic fields has indicated that these previous values were in error by about 1-1/2 per cent. For a more detailed analysis of the calibration procedures and the construction of the apparatus, the reader is referred to Ref. 2.

The diamagnetic susceptibility of artificial graphite is known to be anisotropic. It has been demonstrated, however, that for polycrystalline materials the sum of the susceptibilities measured in three orthogonal directions is independent of the degree of orientation in the material. [3] It is only required that H and the gradient of H be sensibly constant over the volume of the sample. This apparatus meets such requirements and has been used to verify experimentally the uniqueness of this "total susceptibility." Graphites of the same type have been found to differ from sample to sample by less than 1/2 per cent. All values reported herein will be in terms of the total susceptibility.

B. Annealing Techniques

In order to measure the susceptibility in three orthogonal directions,

cubes of graphite were used which could be rotated in the field. The dimensions of the magnet gap limited the size of the samples to less than 1/8-inch. The pulse annealing technique, normally employed in this laboratory,[4] makes use of rectangular specimens, approximately 1 inch long, the samples being heated by a current which passes through them. Due to the cubical shape of the susceptibility samples, such a method was obviously unsatisfactory. The sample was pulse annealed instead in a small vacuum furnace. The furnace is shown in Fig. 1. The furnace heater is a graphite helix of five turns, 6 inches long, and 1/10 inch thick. It is vertically suspended as shown by 1/16 inch copper tubing through which coolant water flows. The graphite helix is attached to the tubing by first electroplating the end of the helix with copper, tinning the copper with soft solder and then soldering the assembly together as shown. An axial radiation shield consisting of eight turns of 5 mil tantalum foil surrounds the helix. It is held in place by strips of nickel which are spot welded to the shield and soft soldered to the tubing. With a power input of approximately 1 kilowatt, a temperature of 2200° C can be reached without difficulty in the center of the helix. Below the helix a Wilson seal is placed, through which an elevator shaft extends up into the heater. The construction of the elevator shaft can be seen in Fig. 1. The shaft itself consists of a hollow stainless steel rod 1/8 inch in outside diameter, with a 1/10 inch bore which is closed at the end by a Vycor metal to glass seal. Two-hole thermocouple tubing passes through the rod and extends 3 inches above it. Attached to this thermocouple tubing is a graphite platform which supports the graphite sample. Five mil platinum and platinum-10 per cent rhodium thermocouple wires were pulled through the Vycor seal and the thermocouple tubing to the sample. The thermocouple junction consists of a butt-weld. A hole was drilled through each sample, and care taken that in each case the butt-weld was located at the center of the sample.

The thermocouple emf was measured using a Leeds and Northrup double range potentiometer. This enables the temperature of the samples to be accurately measured up to 1500° C. Above 1000° C the temperature was also measured using a Leeds and Northrup optical pyrometer. The samples were kept in sight throughout the pulse by observing them through a diagonal mirror which enables one to sight down the helix conveniently. A correction factor for all temperatures obtained using the optical pyrometer was determined empirically as follows: The optical system through which the light traveled

was duplicated, with a standard lamp in place of the furnace. The optical pyrometer temperatures were then compared with the temperature of the standard lamp. The correction ranged as high as +70° C at 2200° C. Making such corrections, temperatures obtained using the optical pyrometer and the thermocouple agreed to within 10° C between 1000° C and 1500° C.

The method of pulse annealing the samples is as follows. The furnace was raised to the desired temperature, with the elevator lifted so that the sample was out of the furnace. To pulse the sample temperature, the elevator was pulled down into the furnace for a period of 3 minutes, and then raised out of the furnace. Temperature readings were made at 15 second intervals. It was found that the sample temperature rose to within 10 degrees of the final temperature in 100 to 120 seconds pulse time over the entire range of pulse temperatures. The temperature dropped sharply after removal of the sample from the furnace. Therefore, the time at temperature was about 1 minute. At lower temperatures the release of stored energy from irradiated graphite caused the temperature of the samples to rise above the temperature of the furnace. However, it was found that by raising or lowering the elevator during this time, the temperature could be maintained to within 5° C. The samples were weighed before and after each pulse, and no change in weight occurred. The vacuum was maintained at a pressure of about 1 micron in the manifold during the pulse by continuous pumping.

Such a pulsing technique was used to pulse a set of irradiated graphite samples step-wise to 2180° C. At the end of each pulse the sample was removed, and the susceptibility was measured at room temperature. The experimental data are presented graphically in Fig. 2, which gives the magnetic susceptibility as a function of annealing temperature. The abscissa is the temperature of the pulse immediately preceding the measurements. The error in pulse temperature measurement is less than 10° C while the magnetic susceptibility measurements are good to $\pm 0.10 \times 10^{-6}$ cgs.

III. CONCLUSIONS

The results of these observations show the existence of two active annealing regions, one between 100° C and 500° C, the other from 1100° C to 1500° C. A similar behavior is also shown by the Hall coefficient during

annealing.[5] All of the graphites show complete recovery with the exception of the 460 mwd sample, which yields a smaller terminal value somewhat outside the experimental error. The failure of complete recovery for this particular sample has been noted for other properties,[3] and indicates that this particular sample is a non-representative piece of material. Another interesting feature of the data is the apparent regression of the susceptibility for the 12.5 mwd/ct sample. The susceptibility reaches a value at about 500° C similar to that of the unirradiated graphites, then falls to a value 2 per cent lower at about 1300° C at which time it begins to recover, reaching the unirradiated value again at about 1900° C. Such behavior is indeed interesting and suggests the desirability of a similar study with a sample with a lower irradiation.

It has been proposed[3] that the diamagnetic susceptibility depends solely upon the number of conduction electrons. It has also been shown, to a first approximation, that the Hall coefficient R is dependent solely upon the number of conduction electrons.[3] From pulse annealing curves of the Hall coefficient, it should therefore be possible to compute the corresponding curves for the susceptibility. Using the Hall coefficient data[5] the susceptibility curves have been calculated using the theoretical formula mentioned above, and these curves are in complete agreement with the observed data of Fig. 2. Thus the present results form a strong argument for the theoretical basis of the calculations and for the hypothesis that the susceptibility is solely dependent upon the number of conduction electrons.

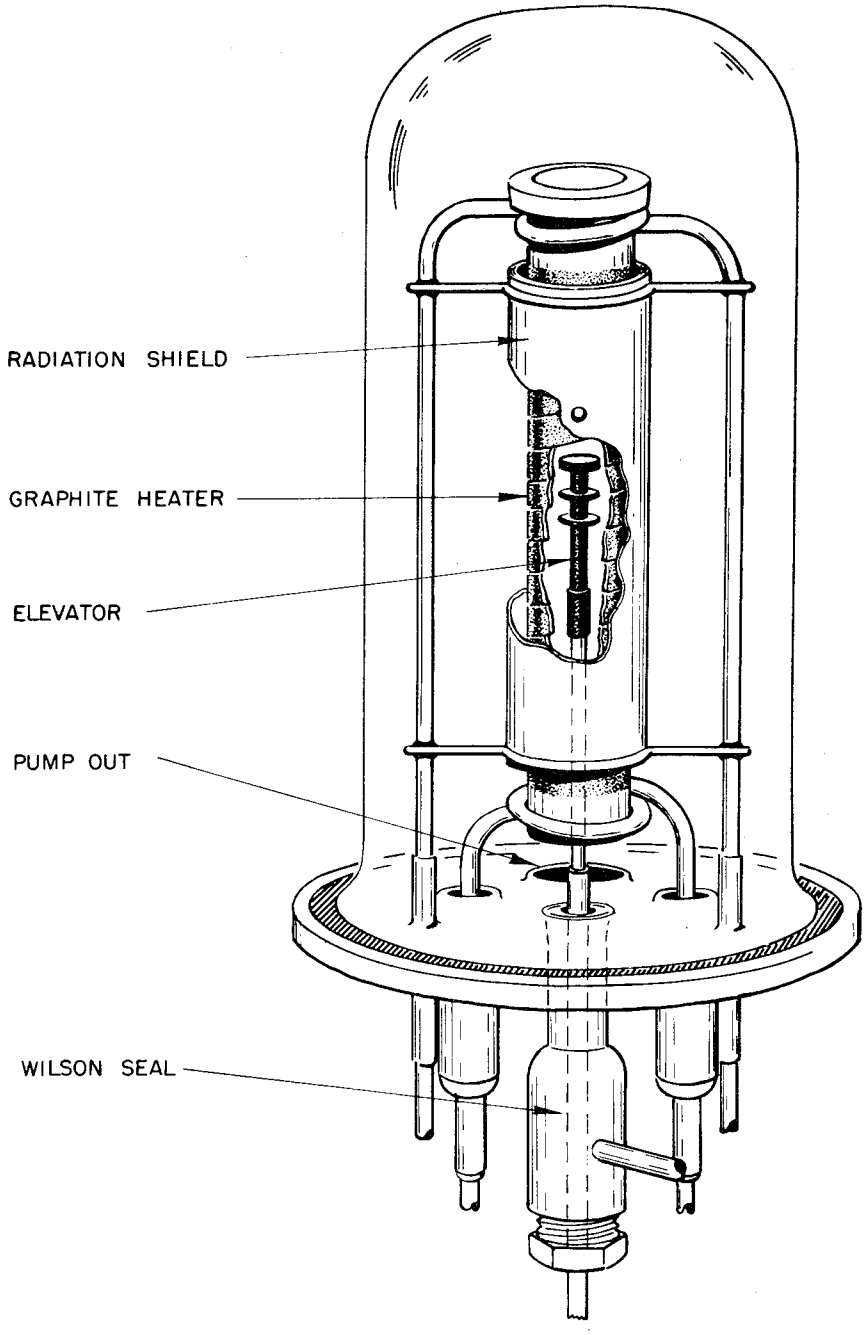

Figure 1. Vacuum Furnace

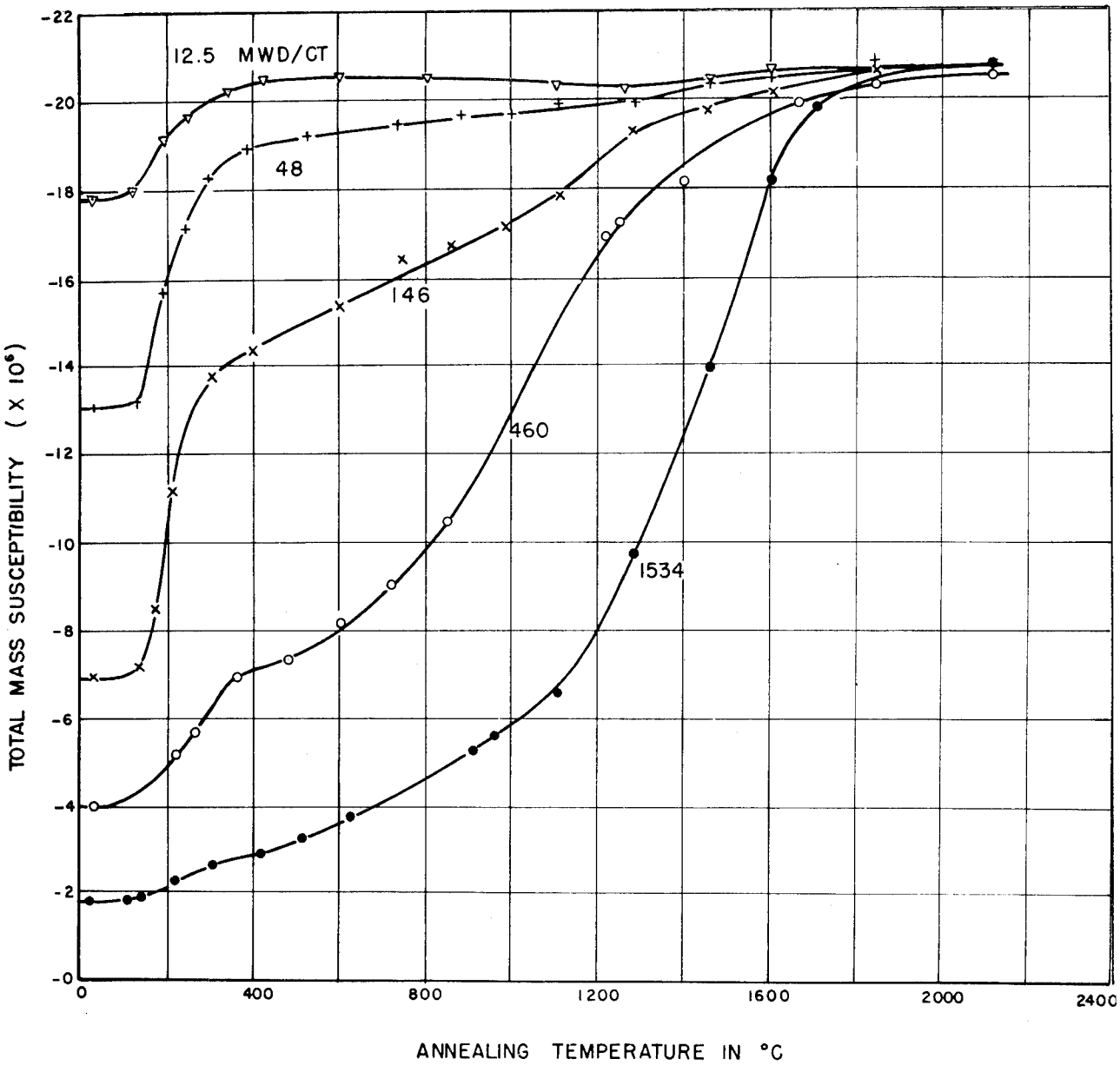

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Figure 2

REFERENCES

1. Donoghue, J. J., and J. D. McClelland, "The Magnetic Susceptibility of Graphite and the Effect of Irradiation on Artificial Graphite," NAA-SR-153, December 18, 1951.

2. Donoghue, J. J., "Magnetic Susceptibility Apparatus," to be published.

3. Eatherly, W. P., "A Theoretical Treatment of Electronic Phenomena in a Two-Dimensional Graphite Lattice," NAA-SR-146, July 25, 1951

4. Parkins, W. E. and J. M. Nuding, "Experimental Technique in Annealing Bombarded Graphite," NAA-SR-15, October 15, 1948.

5. Eatherly, W. P., "Changes in the Hall and Magneto-Resistivity Coefficients During Pulse-Annealing of Irradiated Graphite," NAA-SR-76, September 25, 1951.

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